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Use of lignin biopolymer from industrial waste as bitumen extender for asphalt mixtures

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Abstract

Liquid waste containing lignin, a biopolymer of vegetable origin, is generated from the production of wood hardboards. Because polymers improve the performance of asphalt mixtures, this work studies the possibility of using this industrial waste as a bitumen extender in the production of asphalt mixtures. Thus, asphalt mixtures with 0% (control), 5%, 10%, 20%, and 40% of industrial waste were produced. The bitumen–aggregate adhesion, moisture damage resistance, resilient modulus, permanent deformation resistance, and thermal susceptibility of such mixtures were analysed. Asphalt mixtures with 20% of industrial waste have shown the best moisture damage resistance. As the percentage of industrial waste increased, the thermal susceptibility of the mixture also increased. Therefore, it can be stated that it is appropriate to use this industrial waste containing lignin as a bitumen extender. It can be used in asphalt mixtures for road pavement, mainly by substituting 20% of the bitumen by this liquid waste. It reduces the consumption of bitumen and improves the performance of asphalt mixtures, contributing to the purpose of sustainable construction. The industrial waste was not subjected to any transformation process, thus facilitating its use. The reduction in the use of bitumen in asphalt mixtures by adding this industrial waste contributes to the goals of sustainable development and cleaner production of asphalt mixtures.

Keywords: lignin; biopolymer; industrial waste; asphalt mixtures; bitumen extender

1. Introduction

Polymers are widely used as bitumen modifiers as it has been demonstrated that they can improve the performance of asphalt mixtures. Thus, asphalt mixtures made with polymer-modified bitumen have better rutting performance at high temperatures, higher fatigue resistance, and higher cracking resistance at low temperatures as a consequence of a noticeable reduction in their thermal susceptibility (Brovelli et al., 2015, Ait-Kadi et al., 1996; Hernández et al., 2014). In addition, environmental benefits are associated with the use of modified bitumens owing to the increase in their lifecycle (Vila-Cortavitarte et al., 2018, Brovelli et al., 2014, Brovelli et al., 2013).

Biopolymers, also known as 'green polymers' (Hernández et al., 2014), are organic polymers synthesized by microorganisms, plants, trees, and other biological organisms (Chang et al., 2016). Furthermore, the polymers chemically synthesised from vegetable oils, resins, sugars, and other biological sources could be included in this definition (Hernández et al., 2014). Biopolymers are produced from renewable resources (Chang et al., 2016), which makes them environmentally friendly.

Moreover, biopolymers are harmless and can easily be found in nature (Hataf et al., 2018). In fact, biopolymers can be obtained from several available raw materials (Chang et al., 2016). Lignin is the second most abundant biopolymer on Earth after cellulose (Boerjan et al., 2003), and it is present in all vascular plants (Chávez-Sifontes and Domine, 2013) binding the fibres in wood (Boerjan et al., 2003).

Part of the vegetal biomass containing lignin can be considered as a waste material, which is mainly obtained from the wood, paper, and biofuel industries (Batista et al., 2018) or even from agricultural processes (Chavez-Sifontes and Domine, 2013). Depending on the biomass source and as a consequence of the industrial process used to obtain wood hardboards, paper, etc., various types of lignin with different properties can be obtained. According to the industrial process used, the most common kinds of lignin are the sulphite, kraft, and soda lignin, followed by the organosolv lignin (Chávez-Sifontes and Domine, 2013).

The high abundance of lignin, its cementitious nature, and the fact that lignin and asphalt have some similar chemical compounds, the carbon-rings (aromatics) (Hernández et al., 2014), make this biopolymer particularly interesting for manufacturing asphalt mixtures.

The problem associated with the real overexploitation and limited crude oil resources (Sun et al., 2017) can be mitigated by the use of lignin as a partial substitute of bitumen. Additionally, the use of lignin in bitumen is beneficial for the reduction of CO_2 emissions (van Vliet et al., 2016), because it does not generate any additional CO_2.

Thus, the use of lignin in the manufacture of asphalt mixtures can contribute to the goals of sustainable construction and cleaner production.

Some attempts to use lignin in the production of asphalt mixtures have been reported.

The Federal Highway Administration (FHWA) carried out a study in which the possibility of using wood lignin as a substitute for bitumen and as a bitumen extender in hot and cold asphalt mixtures was analysed (Terrel et al., 1980). As a result, the wood lignin showed promising results as an extender, but not as a bitumen substitute. Nevertheless, this study also concluded that, at that moment, the use of lignin as bitumen extender was not economically feasible.

Sundstrom et al. (1983) stated that asphalt mixtures made with 30% of lignin by weight of bitumen reached the maximum Marshal stability at 6% of binder content.

Williams (2008) used four lignin-containing co-products as bitumen antioxidant, obtaining an improvement of the high temperature properties and a worsening of the low temperature properties of the bitumen. The bioasphalt was derived from lignin-rich corn stover waste. In 2010, a trial bike path was constructed in Iowa replacing 3% of its bitumen by this bioasphalt. The bike path showed adequate performance (Bourzac, 2015).

Pan (2012) stated that lignin could be used as antioxidant for bitumen. However, the lignin temperature must be controlled to avoid lignin aging.

In 2015 (Gosselink, 2016; Wageningen University & Research, 2015), the first test section worldwide was made using bioasphalt from lignin in Zeeland (the Netherlands). The lignin was chemically transformed using fast pyrolysis. The bioasphalt was made by using 50% of lignin. The trial section, 70 m in length, was made with low temperature bioasphalt. After two years, the performance of the road was adequate. Following the same research line, a bicycle path was constructed in 2017 using eight types of lignin.

Xu et al. (2017) demonstrated that lignin-modified bitumen improves the rutting performance at high temperatures and aging resistance of asphalt mixtures. On the contrary, their fatigue life could be reduced.

Xie et al. (2017) also demonstrated that Kraft lignin could be used as bitumen modifier, leading to asphalt mixtures with better performance at high temperature.

Boomika et al. (2017) used plastic and lignin as partial bitumen substitutes. The authors stated that the use of 15% of lignin and 20% of plastic produced efficient results.

Batista et al. (2018) stated that lignin modified bitumen significantly improves the thermal cracking resistance of asphalt mixtures at temperatures as low as −12 °C.

In another research conducted by The Netherlands Organization for Applied Scientific Research (TNO) (Bourzac, 2015; Slaghek et al., 2017; van Vliet et al., 2016), some chemical modifications were made to lignin. As a result, a more hydrophobic lignin was achieved. This modified lignin was blended with bitumen (up to 25% by weight of bitumen). The obtained bitumen presented high resistance to climatic conditions with greater service life. Nevertheless, this bitumen is more expensive than conventional bitumen.

It is interesting to note that most researchers do not use directly lignin waste in bitumen modification for road applications because the untreated lignin does not mix with bitumen (Hernández et al., 2014; Bourzac, 2015).

Other biopolymers have been used in the bitumen industry, such as xantan gum (Tu et al., 2016a), welan gum (Tu et al., 2016b), and starch (Al-Hadidy et al., 2011), but their characteristics make them less suitable than lignin for road pavement applications.

2. Aims and scope

In the present study, industrial waste containing a vegetal lignin biopolymer is used for partial substitution of the bitumen in asphalt mixtures.

Industrial waste from the manufacture of wood hardboards will be used without any transformation, i.e., it will be mixed directly with conventional bitumen. Different percentages of bitumen substitution by industrial waste will be tested, namely, 0% (control mix), 5%, 10%, 20%, and 40% by bitumen weight.

The obtained binder (bitumen + industrial waste) was used to produce AC 22 bin G asphalt mixtures and produce laboratory specimens for testing. The volumetric and mechanical properties of the specimens (indirect tensile strength, water sensitivity, resilient modulus, thermal susceptibility, and permanent deformation resistance) will be analysed.

This work analyses the use of this waste as a partial substitute for bitumen, as a bitumen extender, for application in asphalt mixtures, and the improvement in performance of these asphalt mixtures is also studied.

This work contributes to the purposes of sustainable construction and cleaner production, as industrial waste rich in raw materials will be utilised, without the need for subsequent transformations or production of leftover waste materials.

3. Materials and methods

3.1. Aggregates

A local contractor supplied the aggregates used in this work for the manufacture of asphalt mixtures. The material is Hornfels from a Galician (Spain) quarry (figure 1).

Figure 1. Fraction 4/8 mm of the Hornfels used in this work

It is expected that these aggregates present a poor water resistance because of its mineralogical composition, which is rich in silica (62.30% of SiO_2).

The main properties of the aggregates were analysed according to the Spanish General Technical Specifications for Roads, also known as PG-3 (MFOM, 2015). As can be observed in Table 1, the sand equivalent (SE) complies with the PG-3 for all the traffic categories, and the Los Angeles (LA) abrasion coefficient.

Table 1. Aggregate characterization

Property	Standard	Hornfels	PG-3 Specifications (*)		
			T00-T1	T3-T2	T4
SE (%)	EN 933-8 (AENOR, 2012)	61	≥ 50	≥ 50	≥ 50
LA abrasion (%)	EN 1097-2 (AENOR, 2010)	14.1	≤ 25	≤ 30	-

(*) Traffic category T00 refers to AADHT (Annual Average Daily Heavy Traffic)≥4,000
Traffic category T0 refers to 4,000>AADHT ≥2,000
Traffic category T1 refers to 2,000>AADHT ≥800
Traffic category T2 refers to 800> AADHT ≥200
Traffic category T3 refers to 200>AADHT ≥50
Traffic category T4 refers to AADHT<50

3.2. Bitumen

A 50/70 penetration grade bitumen was chosen to prepare the asphalt mixtures. The bitumen has a penetration of 66x0.1 mm at 25 °C and a softening point of 48 °C.

3.3. Industrial waste containing lignin

The industrial waste used as bitumen extender is a 100% natural product. It is a concentrate of wood extracts, mainly from eucalyptus, from the manufacture of hard fibres boards. As shown in figure 2, the industrial waste is a brown liquid with a faint odour of caramel. Its main properties are listed in Table 2.

Figure 2. Industrial waste containing lignin used in this work

Table 2. Main properties of the industrial waste containing lignin

Property	Value
pH	3.2
Densitity at 75 °C (kg/m^3)	1,155
Viscosity at 80 °C (mPa.s)	14
Total solids (g/L)	444
Volatile solids (g/L)	385
Sulphates (mg/g)	8
Aluminium (Al) (mg/L)	8,197
Calcium (Ca) (mg/L)	4,600
Magnesium (Mg) (mg/L)	5,500
Sodium (Na) (mg/L)	4,725
Silicon (mg/g)	<3
Vanadium (mg/kg)	<0.5
Chlorine (mg/kg)	6,500

The industrial waste has 43.69% of dry matter. This means that 100−43.69=56.31% is water. The main components of this dry matter are 41.46% of sugar (mainly xylose, glucose, galactose, rhamnose, arabinose, and mannose), 23.39% of lignin (16.29% of insoluble Klason lignin and 7.10% of soluble lignin), 13.3% of pectin, 11.8% of polyphenols, 9.05% of mineral matter, and other compounds that appear in low percentages. The high sugar content explains the caramel-like odour, which is particularly noticeable when the compound is heated with the bitumen.

In this work, the industrial waste was added to the hot bitumen (170 °C) in percentages of 0% (control), 5%, 10%, 20%, and 40% and manually blended. Furthermore, a percentage of 60% of industrial waste was added to determine the volumetric properties and stripping potential; however, the poor results obtained caused this percentage to be discarded before the end of the research.

3.4. Asphalt mixture

For this research, the AC 22 base G asphalt mixture for road-pavement base course was chosen. The aggregate gradation limits given by the PG-3 (MFOM, 2015) and the selected aggregate grading curve are shown in figure 3.

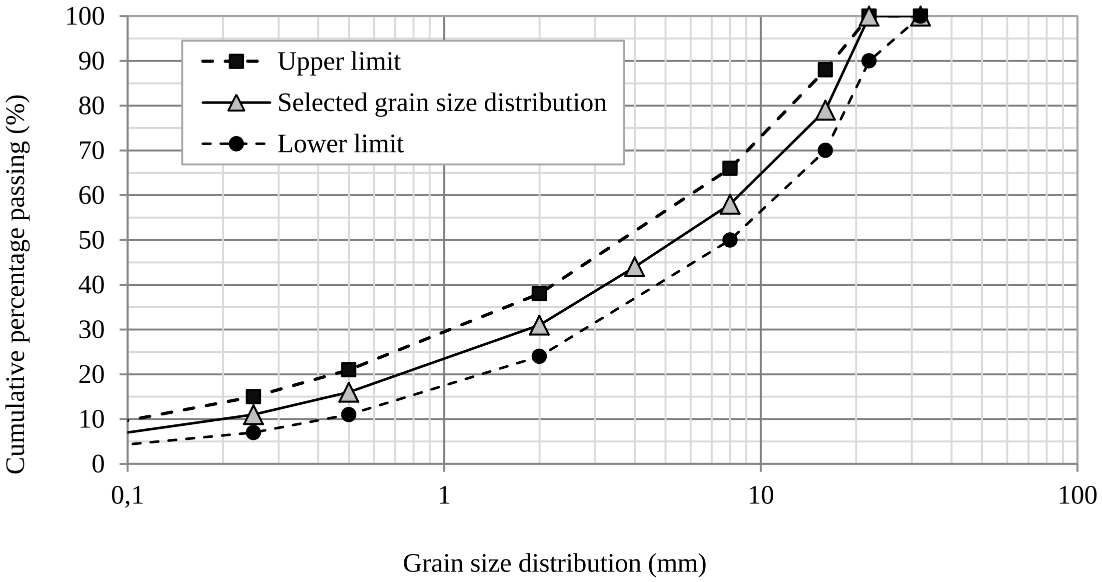

Figure 3. Aggregate gradation curve and standard limits

3.5. Volumetric properties

For the volumetric characterization of the asphalt mixtures, the air voids were determined following UNE-EN 12697-8 (AENOR, 2003) by using the following equation:

$$Va = \frac{\rho_m - \rho_b}{\rho_m} \times 100 \qquad (1)$$

where ρb= bulk specific density and ρm= maximum specific density.

The bulk specific density by saturated surface dry (SSD) was determined on cylindrical Marshall specimens compacted with 75 blows per face according to UNE-EN 12.697-6 (AENOR, 2012c).

The maximum specific density was determined on loose asphalt mixtures according to UNE-EN 12.697-5 (AENOR, 2010b).

According to the PG-3, the minimum bitumen content of the AC 22 base G is 3.9%. Percentages of 3.9%, 4.1%, 4.3%, and 4.5% of binder were used in this work.

3.6. Bitumen and aggregate adhesion

Two qualitative tests, namely the boiling water test and the rolling bottle test have been carried out to analyse whether the incorporation of industrial waste containing lignin improves the adhesion between the aggregate and the binder. Both tests were carried out on loose asphalt mixtures.

3.6.1. Boiling Water Test

The boiling water test has been carried out in accordance with ASTM D3625-96 (2005). In this test, a sample of loose asphalt mixture is introduced in boiling water for 10 min. Afterwards, the bitumen that remains in the water is removed using a towel. Then, the asphalt mixture is removed from the water and dried. The percentage of the aggregate surface that continues coated is visually estimated.

It is not easy to establish a criterion that indicates the suitability of asphalt mixtures in terms of moisture damage resistance (Solaimanian et al., 2003). In the case of the boiling water test, Kennedy et al. (1984) indicate that based on the correlation between tests conducted on the laboratory and field performance, asphalt mixtures that retained less than 70% of the bitumen on the aggregate surface are moisture susceptible. Kim and Coree (2005) indicate that this percentage must be 95%. In any case, results over this percentage cannot guarantee a good field performance owing to other factors contributing to moisture damage resistance (Solaimanian et al., 2003). Thus, this test is useful for initial screening of the asphalt mixtures (Brown et al., 2001) and for identifying problematic materials (Jorgensen, 2002).

3.6.2. Rolling Bottle Method

The rolling bottle test has been carried out in accordance with the UNE-EN 12697-11 (AENOR, 2012) standard. In this test, a sample of loose asphalt mixture is introduced into a bottle containing distilled water and allowed to rotate for 6 h and for 24 h. At the end of these two rotation periods, the percentage of aggregate surface that remains coated with bitumen is visually estimated by two operators.

There are no requirements on the percentage of aggregate surface that should be coated with bitumen after the rotation periods. Therefore, this test is useful for comparison between mixtures produced with different proportions of industrial waste. Particularly, this test is used to analyse the affinity between bitumen and aggregate, strength of the adhesion, and stripping resistance (Solaimanian et al., 2003).

3.7. Moisture damage resistance

To evaluate the water sensitivity of the asphalt mixtures made with 0%, 5%, 10%, 20%, and 40% of industrial waste as bitumen extender, the indirect tensile test was conducted as specified in UNE-EN 12697-12 (AENOR, 2009). This standard indicates that eight cylindrical specimens, compacted with 50 blows per face in the Marshall hammer, must be tested for each binder content.

Four samples will constitute the 'dry set', while the other four will constitute the 'wet set'. The samples of the 'dry set' were kept at room temperature, while the samples of the 'wet set' were saturated in a water bath for 3 days at 40 °C. Then, both sets were left for a minimum of 2 h at 15 °C with the 'dry set' in air and the 'wet set' in water. After this period of time, the tensile strength ratio (TSR) was determined as follows:

$$TSR = \frac{ITS_W}{ITS_D} \times 100 \qquad (2)$$

where TSR = the tensile strength ratio (%), ITS_W = the average tensile strength of the 'wet set' (MPa), and ITS_D = the average tensile strength of the 'dry set' (MPa).

The PG-3 (MFOM, 2015) requires a TSR≥80% to accept an AC 22 base G in terms of moisture damage resistance.

3.8. Resilient modulus and thermal susceptibility

The resilient modulus of the asphalt mixtures was calculated following the Annex C of UNE-EN 12697-26 (AENOR, 2012). In this test, compressive repeated haversine loads were applied in a vertical diametral plane of cylindrical specimens compacted with 75 blows per face with the Marshall hammer. The repetition period of the impulse is 3±0.1 s and the rise time is 124±4 ms. The maximum load must lead to a maximum horizontal strain of 0.005% of the specimen diameter. In this test, 10 conditioning pulse cycles were followed by 5 load pulse cycles. Then, the resilient modulus is determined as follows:

$$M_R = \frac{F \times (\nu + 0{,}27)}{z \times h} \qquad (3)$$

where MR=resilient modulus (MPa), F=maximum applied load (N), z=horizontal deformation (mm), h=sample thickness (mm), and ν=Poisson's ratio (assuming a Poisson's ratio of 0.35 for the different temperatures according to UNE-EN 12697-26 (AENOR, 2012).

The test was conducted at different service temperatures with the purpose of analysing the influence of the industrial waste on the thermal susceptibility of the mixtures.

The thermal susceptibility may be defined as the resistance of the binder to change its resilient modulus and viscosity with temperature (Cong et al., 2012). The lower temperature dependence the asphalt mixture has, the less thermal susceptible the binder is (Cong et al., 2012).

In order to avoid problems of non-linearity, the temperatures have been chosen below 40 °C. Particularly, the samples have been tested at temperatures of 2 °C, 10 °C, 20 °C, and 30 °C.

Furthermore, the mixtures have been tested after an initial curing time of 14 to 42 days (as stated in the standard) and before a period of 6 months with the aim of analysing the effect of ageing time in the thermal susceptibility.

3.9. Permanent deformation resistance

A repeated load axial test without confinement was conducted in order to evaluate the resistance to permanent deformation. The Cooper NU 14 tester machine was used to conduct this test. The British Standard DD 226:1996 (BSi, 1996) describes this test as follows: each cylindrical Marshall specimen must be held at 30 °C in a climate chamber for a minimum of 4 h, and then preloaded for 600±6 s at 10 kPa of axial stress with the load platens. After this time, each specimen was subjected to 1,800 load cycles. In order to simulate loading in the field, square repeated load pulses were selected. The width pulse and the rest period were of 1 s, while the stress level was of 100±2 kPa.

The axial permanent strain was calculated as follows:

$$\varepsilon_{d(n,T)} = \frac{\Delta h}{h_0} \text{x}100 \quad (3)$$

where $\varepsilon_{d(n,\ T)}$ = the axial permanent strain (%) after n load applications at temperature T in °C, Δh = the axial permanent deformation (mm), and h_0 = the initial distance between the two load platens (mm).

A group of specimens has been tested just after it was manufactured, whereas the other group of specimens was tested after a period of 6 months with the aim of analysing the effect of ageing time in the permanent deformation resistance.

4. Results and Discussion

4.1. Volumetric properties

Figure 4 shows the air void content (Va) versus binder content of the asphalt mixtures, for each industrial waste percentage. This figure also shows the Va limits required by the PG-3.

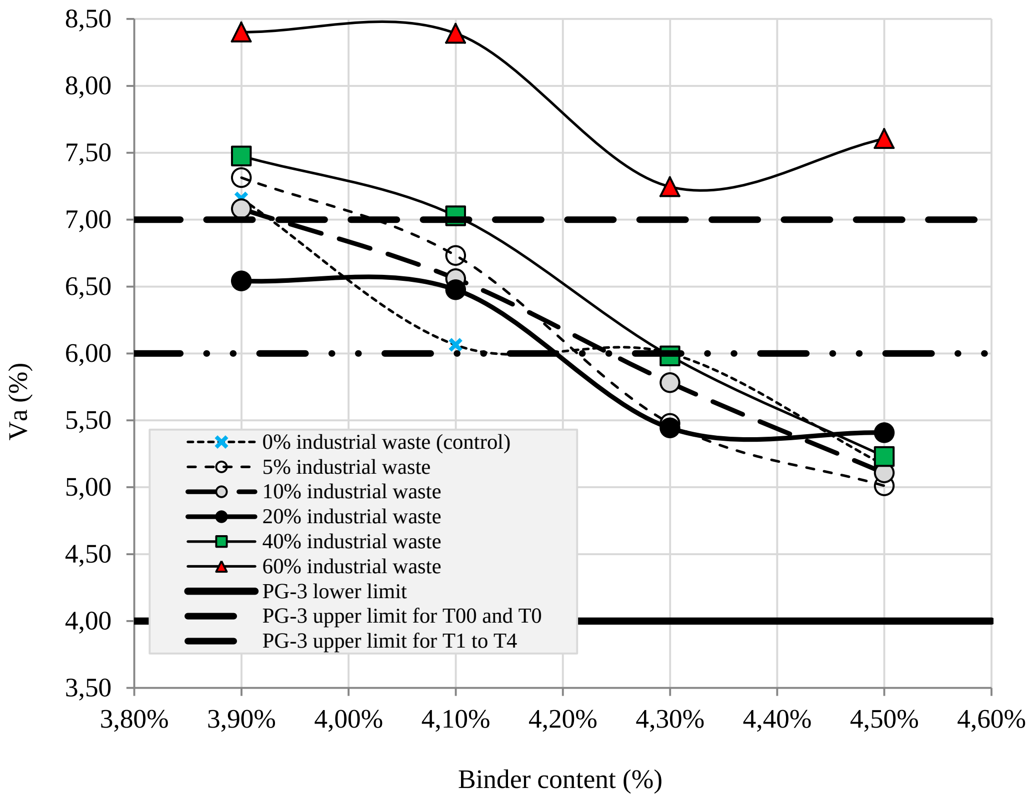

Figure 4. Air void content versus bitumen content for different percentages of industrial waste

The results presented in figure 4 show that the Va decreases as the binder content increase, but mixtures made with 60% of industrial waste show an irregular behaviour. Probably, the high water content introduced in the mixture is mainly responsible for this performance.

Figure 4 shows that there is not a clear trend between Va and industrial waste content. Nevertheless, mixtures made with 40% of industrial waste generally present a Va higher than that of mixtures made with lower waste percentages. This trend is even clearer when analysing the asphalt mixtures made with 60% of industrial waste. In this case, Va is much higher than the one obtained for mixtures with 0% to 40% of waste. Clearly, when a very high amount of industrial waste is used, the compaction is more difficult. Thus, it must be said that some crystallized particles can be appreciated at first sight when 40% and 60% of industrial waste were used. When 20% or less waste is used, this phenomenon is not appreciated. Probably, when higher amounts of waste are used, some coalescence tend to occur, leading to these crystallised particles, and thus, to a heterogeneous structure. This could also explain the irregular performance of mixtures made with bitumen blended with 60% of industrial waste.

Moreover, as observed in figure 4, mixtures made with 0% (control mixture), 5%, 10%, 20%, and 40% of industrial waste containing lignin biopolymer and with 4.30% and 4.50% of binder content comply with the PG-3 for all the heavy traffic categories. Mixtures made with 0%, 5%, and 10% of industrial waste and binder content of 4.10% comply with the PG-3 for T1 to T4 heavy traffic categories. The same occurs with mixtures made with 20% of industrial waste and 3.90% and 4.10% of binder content. Mixtures made with 60% of industrial waste do not comply with the PG-3 for any of the heavy traffic categories.

It is interesting to note that when industrial waste is blended with bitumen, some foaming occurs owing to the sudden warming of the water included in this waste. As the industrial waste percentage increases, the foaming is more pronounced in such a way that for 40% and 60% of industrial waste, it is difficult to operate the blend owing to the high foam formation. This fact can be observed in figure 5, which shows 20% of industrial waste blended with bitumen. At the beginning (figure 5a), the bitumen shows its usual appearance, but after a few seconds (figure 5b), foaming leads to bubble formation inside the bitumen, which is clearly noticeable in its surface. Moreover, a high volume expansion of the bitumen is produced when foaming occurs. The arrows included in figure 5 show this increase in volume.

Figure 5. Bitumen with 20% of industrial waste: a) at the beginning of the mixing process and b) after 5 s of the beginning of the mixing process

The irregular behaviour of the mixtures made with 60% of industrial waste, together with the large Va and the difficulty in using the binder made with bitumen and 60% of waste, caused these mixtures to be discarded.

4.2. Bitumen and aggregate adhesion

Figure 6 shows the percentage of aggregate surface that remains coated after 24 h of rolling time in the rolling bottle test and after 24 h of drying time in the boiling water test.

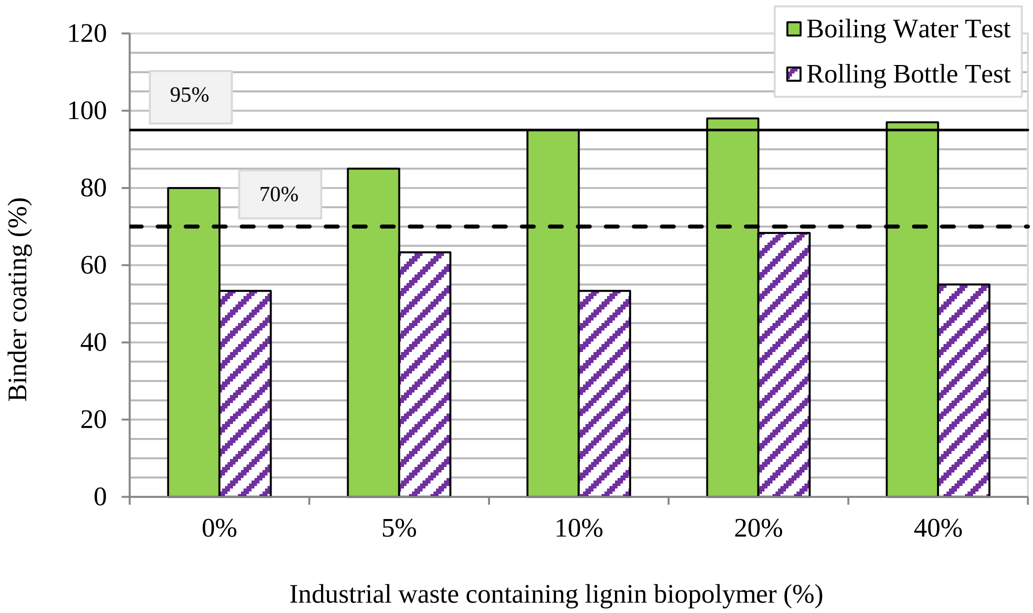

Figure 6. Bitumen-aggregate adhesion results after 24 h

Figure 6 shows that all tested mixtures lead to boiling water test results over 70%, which is the minimum required by some researchers to accept the material. However, only the mixtures made with 10% to 40% of industrial waste lead to boiling water test results over 95%, which is the minimum required by other researchers.

Both tests show that the best binder coating always occurs for mixtures with 20% of industrial waste. In general, for industrial waste percentages of 0% to 20%, as the industrial waste percentage increases, the percentage of aggregate surface that remains coated with the binder also increases. The only exception of this trend is the result of the rolling bottle test for the blend with 10% of industrial waste, which is clearly due to the typical dispersions of these tests. When the highest amount of industrial waste is used (40%), the coated surface is reduced. As said before, the high water content and the formation of crystallised particles could also explain this performance.

Therefore, the use of industrial waste containing lignin biopolymer as extender improves the affinity between bitumen and aggregate when using percentages up to 20%. As said before, higher percentages introduce high water amounts and lead to the formation of crystallised particles. Both seem to affect the affinity between aggregates and binder. At lower percentages, the effect of the lignin included in the industrial waste and the foaming effect of the water lead to better affinity between the aggregates and the binder. It is possible that the foam formation as a consequence of the industrial-waste water evaporation improves the aggregate–binder mixing, thus improving the aggregate–bitumen adhesion.

Nevertheless, the water sensitivity of the mixture is conditioned by factors different from the binder–aggregate affinity. In this regard, it is necessary to analyse the water resistance of the asphalt mixtures on compacted specimens, as described in the following section.

4.3. Moisture damage resistance

Figure 7 shows the TSR versus binder content for mixtures made with 0%, 5%, 10%, 20%, and 40% of industrial waste containing lignin.

For mixtures made with 0% of industrial waste (control mixture), only 4.5% of binder content achieves the minimum TSR of 80%, required by the PG-3 for AC 22 base G. Thus, the optimum binder content for the control mixture is 4.5%. This binder content leads to mixtures that can be used in all heavy traffic categories, as said in section 3.1.

For mixtures made with 5%, 10%, and 40% of industrial waste, none of the tested binder contents led to mixtures with adequate water sensitivity.

The mixture made with 20% of industrial waste achieved the minimum TSR with 4.1% of binder content. Thus, the optimum binder content for this mixture is 4.1%. This bitumen percentage leads to mixtures that can be used in heavy traffic categories T1 to T4, as said in section 3.1.

It means that using a 20% of industrial waste as bitumen extender requires less bitumen content than the control mixture to achieve an adequate water resistance. Nevertheless, the mixtures should be used for heavy traffic categories T1 to T4 (medium and low traffic roads).

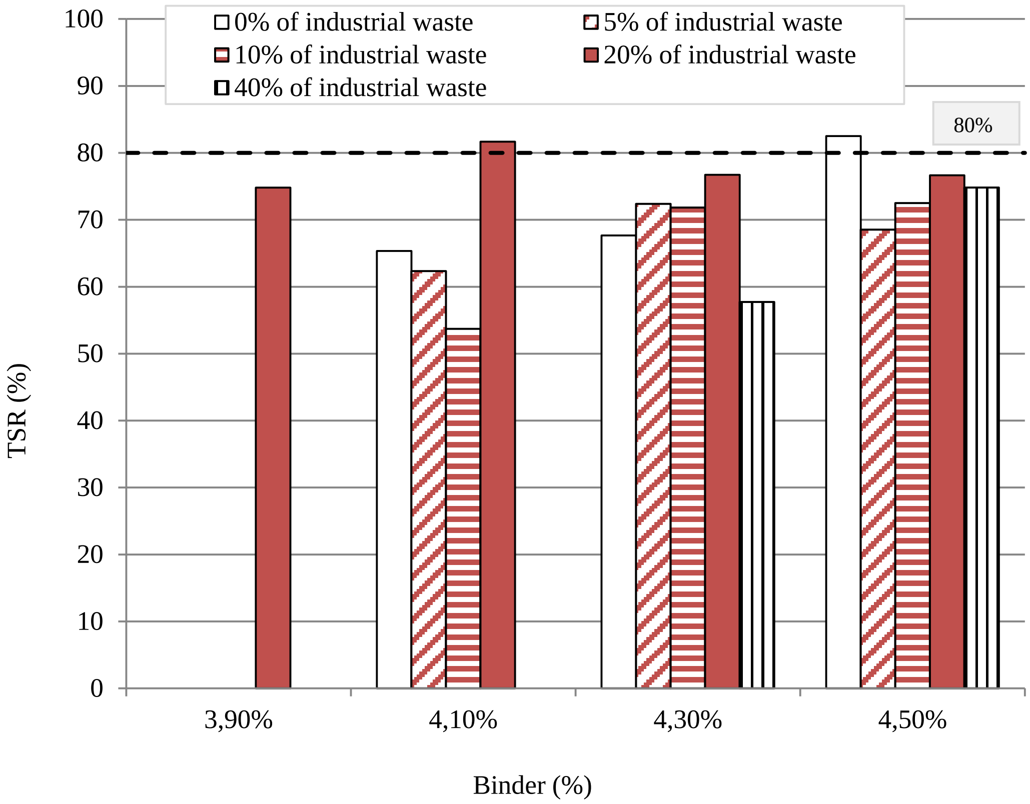

Figure 7. TSR vs binder content for mixtures with 0%, 5%, 10%, 20%, and 40% of industrial waste

Figure 8 shows the indirect tensile strength of the dry and wet sets for mixtures made with 0%, 5%, 10%, 20%, and 40% of industrial waste containing lignin. This figure clearly shows that mixtures made with 20% of industrial waste present dry (figure 8a) and wet (figure 8b) indirect tensile strengths higher than the control mixture, for all the tested binder contents.

It is possible to observe that mixtures made with 20% of industrial waste at their optimum bitumen content (4.1%) display higher dry indirect tensile strength than the control mixture at its optimum bitumen content (4.5%). Regarding the wet indirect tensile strength, for mixtures made with 20% of industrial waste it is slightly lower, but very similar. Thus, there is not a loss of indirect tensile strength when using 20% of industrial waste at the optimum bitumen content.

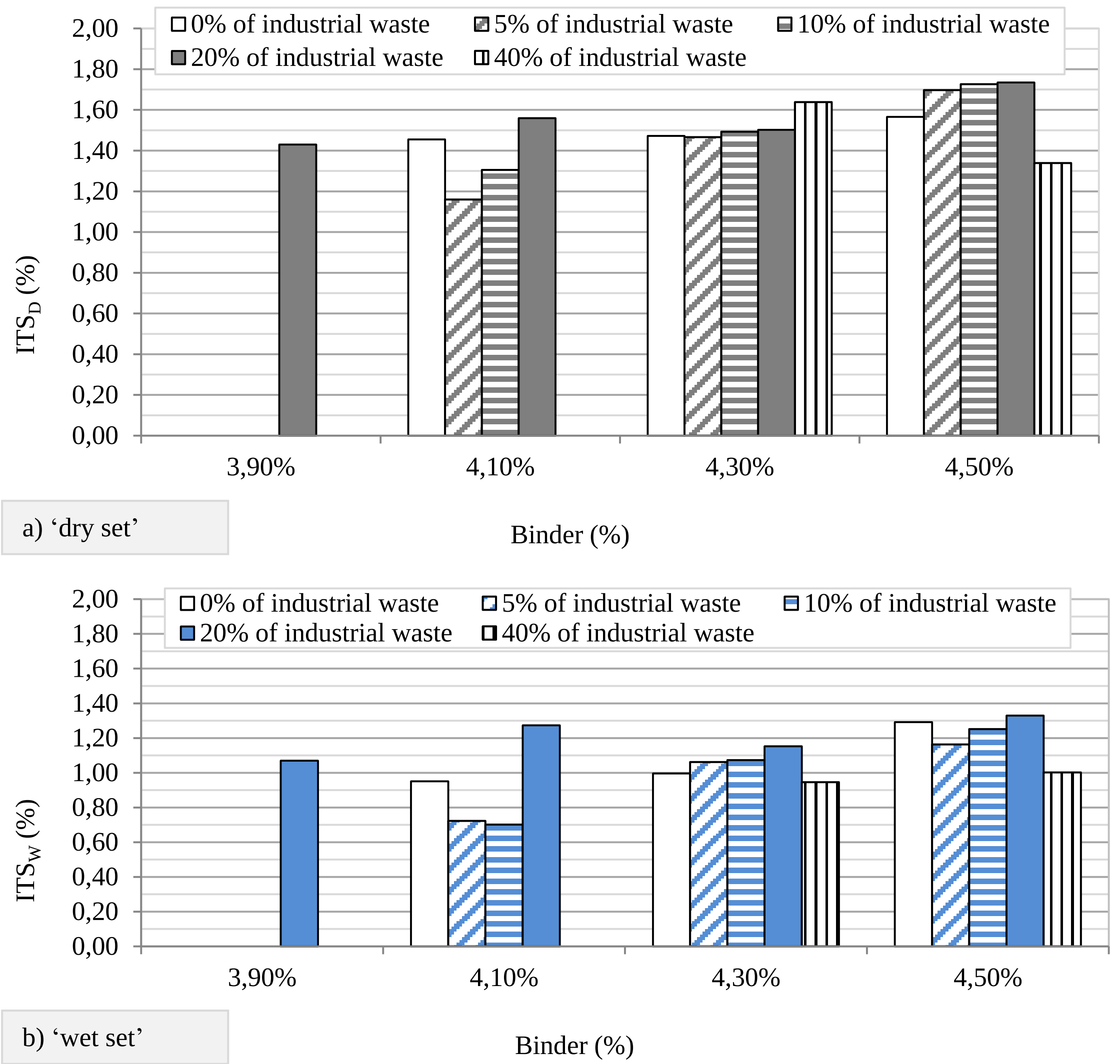

Figure 8. Indirect tensile strength versus binder content for mixtures made with 0%, 5%, 10%, 20%, and 40% of industrial waste: a) 'dry set' and b) 'wet set'

These results allow to state that the percentage of 20% of industrial waste as bitumen extender seems to be the most adequate for the production of asphalt mixtures. It improves the affinity between the aggregate and the binder and reduces bitumen consumption in terms of water sensitivity for low and medium traffic roads.

Industrial waste percentages of 10% or less seem not to have enough lignin to appreciate its effect on the asphalt mixtures. In these cases, the negative effect of the water content appears to be more noticeable than the effect of the lignin. In contrast, industrial waste percentages of 40% or more introduce heterogeneities in the asphalt mixtures owing to their high water content and lead to the formation of crystallised particles that negatively affect the performance of the asphalt mixtures. Thus, 20% is the optimum amount of industrial waste containing lignin biopolymer.

4.4. Resilient modulus and thermal susceptibility

Figure 9 shows the resilient modulus for an AC 22 base G made without industrial waste at its optimum bitumen content (4.5%) and for an AC 22 base G made with 20% of industrial waste, also at its optimum bitumen content (4.1%). The resilient modulus was obtained at 2 °C, 10 °C, 20 °C, and 30 °C.

Figure 9 clearly shows that as the temperature increases, the resilient modulus decreases owing to the thermoplastic nature of the binder.

In figure 9a, the differences between the control mixture and that with 20% of industrial waste can be appreciated. Clearly, at low temperatures, the resilient modulus of the control mixture is higher than that of the mixture made with 20% of industrial waste. The opposite occurs at the highest temperatures. Thus, the thermal susceptibility of the mixtures made with 20% of industrial waste is lower than that of the control mixture.

Low thermal susceptibility controls permanent deformation at high temperatures and avoids fracture at low temperatures (Zapién-Castillo et al., 2016). Thus, the performance of the asphalt mixture made with 20% of industrial waste will be higher, leading to higher fatigue resistance at low temperatures and higher resistance to permanent deformation at high temperatures.

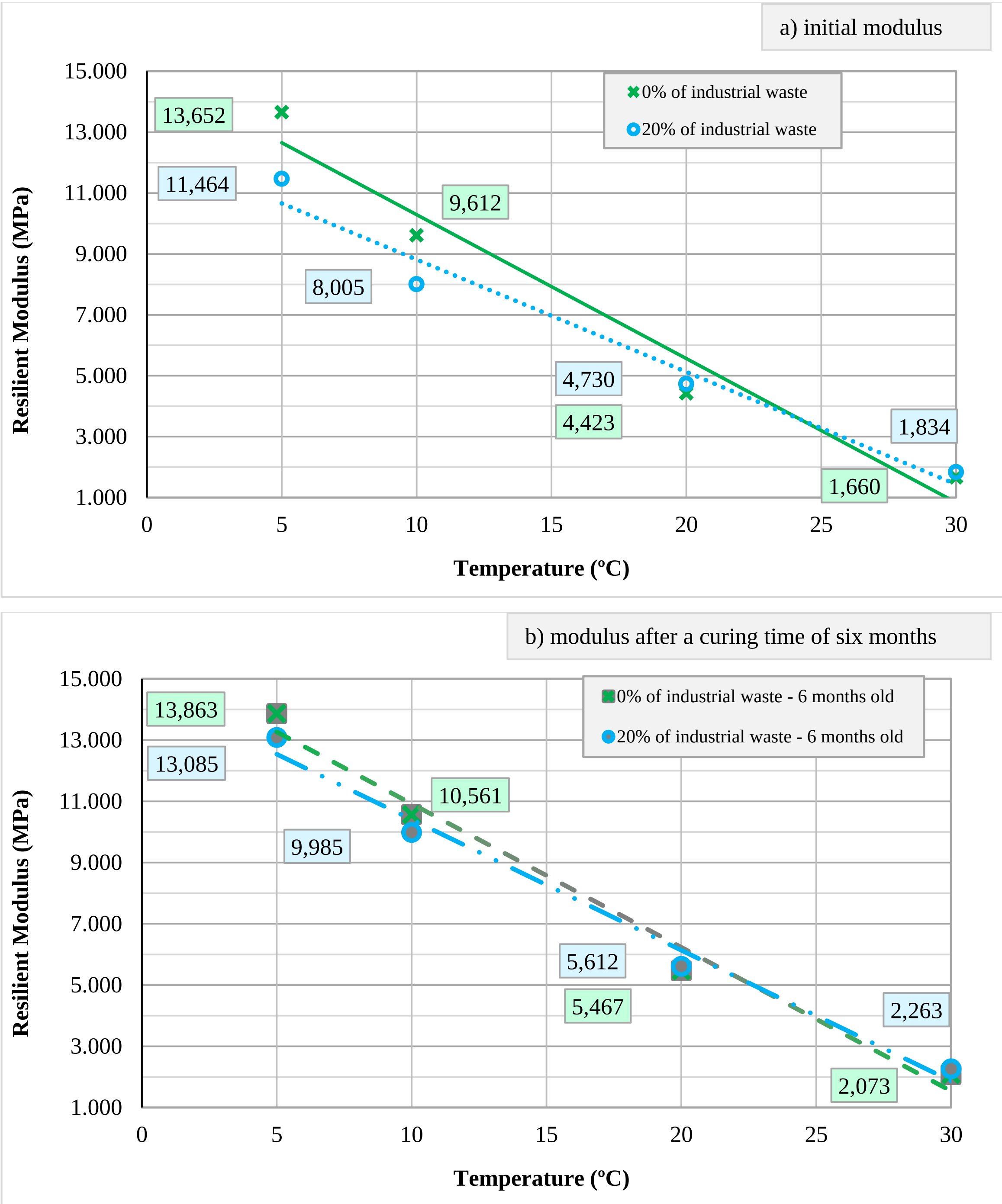

Figure 9. Resilient modulus at different temperatures: a) initial modulus and b) modulus after a curing time of six months

Figure 9b also confirms this conclusion. Asphalt mixtures made with 20% of industrial waste containing lignin lead to low thermal susceptibility. Nevertheless, it must be taken into account that after six months of curing time the resilient moduli are higher owing to ageing of the mixture. Furthermore, the thermal susceptibility of mixtures made with 20% of industrial waste is not as low as that at the initial moment. That is, the differences between the control mixture and the mixture made with 20% of industrial waste are less pronounced as time elapses.

4.5. Resistance to the permanent deformation

Figure 10 shows the axial permanent deformation vs. number of load cycles for an AC 22 base G for the control mixture at its optimum bitumen content (4.5%) and for an AC 22 base G made with 20% of industrial waste at its optimum bitumen content (4.1%). The permanent deformation resistance was obtained at 30 °C.

This figure includes the results of the initial permanent deformation and the results for mixtures cured during 6 months at room temperature.

Because no specifications have been included in the PG-3 for repeated load axial test results, this test is useful for comparison.

Figure 10 clearly shows that the mixture made with 20% of industrial waste presents lower permanent deformation than the control mixture (0%) at an initial moment. These results are coherent with those obtained in figure 9. In this way, at the initial moment, the stiffness at 30 °C is higher for mixtures made with 20% of industrial waste than that for the control mixture. According to this, the deformation will be lower in the stiffer mixture (20% of industrial waste).

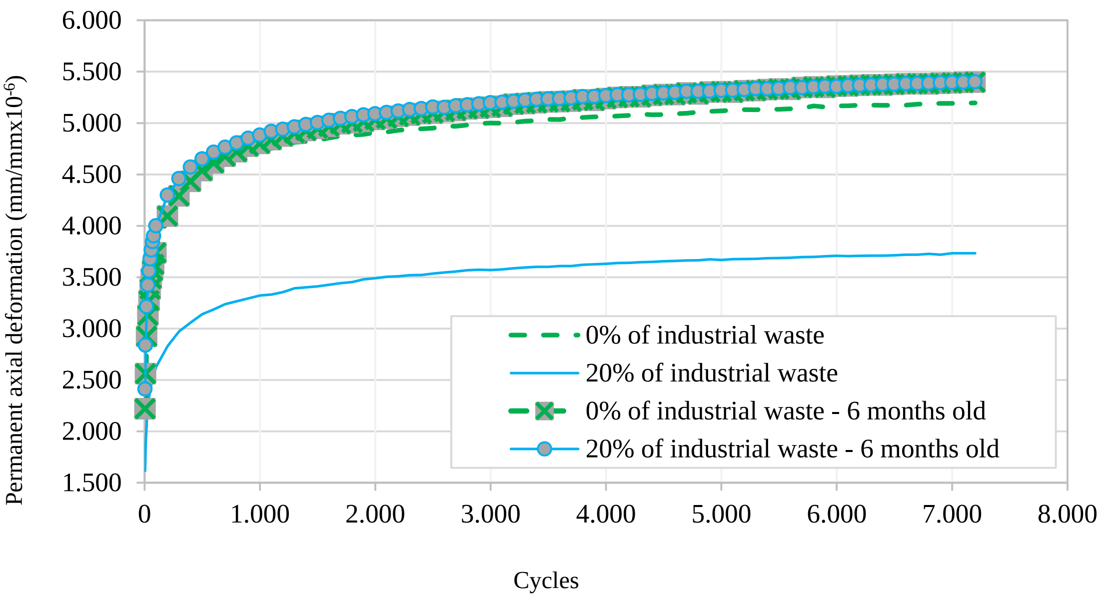

Figure 10. Repeated load axial test results

Nevertheless, the differences between mixtures made with 0% and 20% of industrial waste are not noticeable when 6 months of curing time have passed. Again, the results are coherent with those obtained in figure 9, because after 6 months of curing time the stiffness at 30 °C is similar for both mixtures.

5. Conclusions

In this research, the feasibility of using industrial waste rich in lignin vegetable biopolymer as a bitumen extender for the production of asphalt mixtures is analysed. The asphalt mixture type AC 22 base G for base course of road pavements was chosen for this work. The bitumen was mixed with 0% (control), 5%, 10%, 20%, 40%, and 60% of industrial waste. The following conclusions were drawn:

- The industrial waste was used without any transformation, thus contributing to economical and cleaner production and to the goal of sustainable construction.
- It is interesting to note that, when the industrial waste is blended with the bitumen, some foaming occurs owing to the sudden warming of the water in contact with the hot bitumen. As the percentage of lignin-containing industrial waste increases, the foaming is more pronounced. Thus, for 40% and 60% of industrial waste, it is difficult to operate with the blend owing to the high degree of foam formation. Nevertheless, for lower bitumen contents (0%, 5%, 10%, and 20%), foaming facilitates the mixing operation and leads to a better aggregate–bitumen adhesion.
- When a large amount of industrial waste was used (40% and 60%), some crystallised particles could be observed.
- For all these reasons, the mixture with 60% of industrial waste showed excessive air-void content and was discarded.
- The affinity test showed that the use of industrial waste containing lignin biopolymer as extender, in percentages up to 20%, improves the affinity between the bitumen and the aggregate.
- The water resistance test showed that the 0% (control) and 20% of industrial waste mixtures displayed the best moisture damage resistance. Thus, the TSR for the control mixture was 82.51% and that for the mixture produced with 20% of industrial waste was 81.67%. In both cases, the TSR complied with the Spanish specifications.
- Nevertheless, these results were achieved for 4.5% and a 4.1% of bitumen content respectively. That is, the control mixture required a binder (bitumen+industrial waste) content 9.8% higher than the binder content of the mixture made with 20% of industrial waste in place of the bitumen. That is, the control mixture consumes 37.2% more bitumen than the mixture made using 20% of industrial waste in place of the bitumen.

- However, the control mixture is adequate for all types of heavy traffic while the mixture made with 20% of industrial waste is adequate only for medium and low traffic roads.
- At low temperatures, the resilient modulus of the control mixture (0% of industrial waste and 4.5% of binder content) is higher than the resilient modulus of the mixture made with 20% of industrial waste (and 4.1% of binder content). The opposite occurs at the highest temperatures. Thus, the thermal susceptibility of the mixtures made with 20% of industrial waste is lower than that of the control mixture. That is, the control mixture presents higher fatigue resistance at low temperatures and higher resistance to permanent deformation at high temperatures. Nevertheless, the thermal susceptibility differences between the control mixture and the mixture made with 20% of industrial waste are less pronounced as time elapses.
- The mixtures made with 20% of industrial waste presented lower permanent deformation than the control mixture (0%) at an initial moment. However, after 6 months of curing time, the differences between the mixtures made with 0% and 20% of industrial waste are not noticeable.
- For all these reasons, it can be stated that the mixtures made with 20% of industrial waste showed the best results for medium and low traffic roads (T1 to T4), taking into account the affinity, water resistance, thermal susceptibility, resistance to permanent deformation, sustainability, and economy.
- Nevertheless, the effect of the industrial waste rich in lignin biopolymer in terms of resilient modulus and permanent deformation of the asphalt mixture is particularly noticeable during the first months of the useful life of the mixture.

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